

# Dynamics of Inter-Landau-Level Excitations of a Two-Dimensional Electron Gas in the Quantum Hall Regime

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The femtosecond inter-Landau-level dynamics of a two-dimensional electron gas in a large magnetic field is investigated by degenerate four-wave mixing on modulation doped quantum wells. We observe a large transfer of oscillator strength to the lowest Landau level, and unusual dynamics due to Coulomb correlation. We interpret the effects using a model based on shakeup of the electron gas.

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Effects of Coulomb correlation manifest themselves in almost all transport and optical properties of semiconductors [1]. They dominate, in particular, the physics of electron-hole ( $e$ - $h$ ) pairs photoexcited near the fundamental optical band gap [2]. In undoped semiconductors, the lowest electronic excitations are high energy and can thus adjust almost instantaneously to the dynamics of the low energy, near-band-gap carriers [3]. Thus, the photoexcited  $e$ - $h$  pairs behave as quasiparticles with mutual interactions, while the ground state, except for providing the band structure and dielectric screening, can be considered as rigid [4]. Then the only Coulomb correlations that matter are dynamically generated by the optical excitation [5]. Correlation effects in photoexcited undoped semiconductors have been extensively investigated over the past decade [2]. Time resolved nonlinear spectroscopy experiments have given direct evidence of four-particle and higher Coulomb correlation effects that require theoretical treatment beyond the mean-field, or random phase approximation (RPA) [6].

Perhaps the most widely used theoretical approach for describing these results is the dynamically controlled truncation scheme (DCTS) [7,8]. In this theory, the response of the semiconductor is expanded in terms of the number of  $e$ - $h$  pairs, and consistently truncated. This can be accomplished because of the correspondence between the number of  $e$ - $h$  pairs in the system and the sequence of photon absorption and emission. In this way, it is possible to systematically include all correlations which contribute to a specified order in the applied field. However, if carriers are present in the system before excitation, this correspondence breaks down, and the DCTS fails. This is the case, for instance, in modulation doped quantum wells (MDQWs), where a strongly correlated two-dimensional electron gas (2DEG) exists in the sample and can react to photons and photoexcited carriers. Clearly, the almost un-

explored dynamics induced by the presence of low energy excitations able to interact with photoexcited  $e$ - $h$  pairs raises formidable theoretical difficulties. Previous efforts have developed a formalism able to handle the case where correlation with an electron Fermi sea dominates the coherent optical response [9]. The picture becomes more complicated when a magnetic field is applied, as illustrated by the literature on the quantum Hall effects [10]. While the excitation spectrum has been predicted [11], few experiments [12] have successfully accessed this information. Charged excitons [13] have been observed in photoluminescence experiments but are not expected to contribute significantly to the coherent response, where the total oscillator strength of the intrinsic excitations dominates the signal. Recently, the first coherent wave mixing study of electronic dephasing in the presence of a 2DEG in the QH regime was reported [14].

We present here the first investigation of the dynamics of the 2DEG inter-LL (Landau level) excitations using time resolved nonlinear spectroscopy. We observe strong, time-dependent Coulomb coupling between the LLs induced by the 2DEG, which enhances the LL0 signal. The latter shows unusual behavior as a function of time delay, which cannot be understood in terms of the RPA. These results are compared directly with measurements on undoped quantum wells (QWs). We also introduce a new theoretical approach that treats the interactions of the magnetoexcitons with the 2DEG excitations and qualitatively accounts for the most salient experimental results.

The samples studied here are multiple period QWs, antireflection coated and mounted on sapphire windows for transmission measurements. We performed measurements on MDQWs whose active region consists of 10 periods of a 12 nm GaAs well and a 42 nm  $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$  barrier, the central 12 nm doped with Si. The carrier density under illumination is  $n = 2.1 \times 10^{11} \text{ cm}^{-2}$ .

The sample has a low temperature mobility of  $\mu \approx 10^5 \text{ cm}^2/\text{Vs}$ . In the measurements discussed here, the total number of carriers excited by the laser was kept below  $2 \times 10^{10} \text{ cm}^{-2}$ , or  $n/10$ . Comparison measurements were made with undoped samples with similar well and barrier sizes. We used two criteria for these comparisons by tuning the laser (i) to excite the same number of  $e-h$  pairs into each LL with a given laser pulse, or (ii) to produce the same four-wave mixing (FWM) signal in the nonlinear susceptibility approximation [15]. The effects reported here were observed for both conditions. The samples were immersed in superfluid helium in a magneto-optic cryostat, at a temperature of 1.7 K. We performed spectrally resolved four-wave mixing (SR-FWM) experiments using two equal intensity laser beams from a mode-locked Ti:sapphire laser with a pulse duration of  $\tau \approx 150 \text{ fs}$  [14]. The laser was tuned to excite varying proportions of the lowest LL (LL0) and the next highest LL (LL1), the beams in direction  $\mathbf{k}_2$  and  $\mathbf{k}_1$  were  $\sigma^+$  circularly polarized, and separated by a time delay  $\Delta t$ . The FWM signal in direction  $\mathbf{k}_s = 2\mathbf{k}_2 - \mathbf{k}_1$  was spectrally dispersed using a 0.75 m focal length spectrometer, and measured with a CCD camera or a photomultiplier tube.

Typical SR-FWM signals,  $S_{\text{SR}}(\Delta t, \omega)$ , for the MDQWs and undoped QWs samples are shown in Fig. 1, with the laser tuned to excite both LL0 and LL1 equally (the laser and absorption spectra are projected on the back panels). Several unusual features are immediately apparent in the signal from the doped QWs,  $S_{\text{SR}}^{\text{doped}}(\Delta t, \omega)$  [Fig. 1(a)]. The most striking is that despite an equal excitation of both LLs, the MDQWs show a LL0 signal which is 35 times larger than the LL1 signal. Measurements performed on the undoped QWs,  $S_{\text{SR}}^{\text{undoped}}(\Delta t, \omega)$  [Fig. 1(b)], show almost equal emission from both LLs, in proportion to the excitation as expected. In addition to the transfer of oscillator strength to LL0,  $S_{\text{SR}}^{\text{doped}}(\Delta t, \omega)$  also shows a very unique dependence on  $\Delta t$ . Although we see emission almost entirely from LL0, the signal has very pronounced beats as a function of  $\Delta t$ , with a period given by the inverse of the energy difference between LL0 and LL1. Such strong beating in  $\Delta t$  from only a single emission energy is astonishing. Comparing this to the signal from the undoped QWs, we see that  $S_{\text{SR}}^{\text{undoped}}(\Delta t, \omega)$  also shows beats, but as expected for two peaks of comparable height.

The picture becomes only more intriguing when we tune the laser frequency to excite mostly into LL1, with only the tail of the laser pulse exciting LL0. The inset of Fig. 2 shows  $S_{\text{SR}}(\Delta t = 0, \omega)$ , the spectra for  $\Delta t = 0$ , for both samples under these excitation conditions. It is clear again that the signal from LL0 is greatly enhanced relative to LL1 in the MDQWs. In the undoped sample, there is almost no signal from LL0, as expected from the excitation (60:1 excitation of LL1 over LL0), while in the doped sample the LL0 signal is comparable to the LL1 signal. We estimate the enhancement of the LL0 signal by comparing the relative emission of the two LLs. We define the emission ratio  $R$  as  $R = (S_m^{\text{LL0}}/N_{\text{LL0}})/(S_m^{\text{LL1}}/N_{\text{LL1}})$ ,

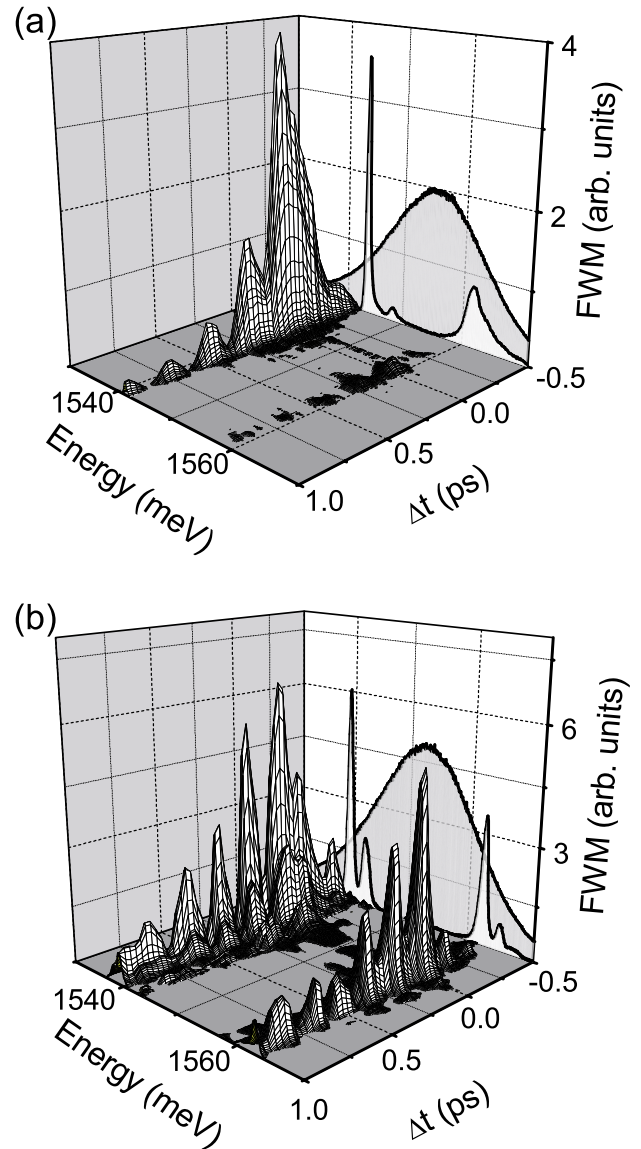


FIG. 1. Spectrally resolved FWM signal at  $B = 8 \text{ T}$  for laser exciting an equal number of  $e-h$  pairs into both Landau levels 0 and 1, for (a) the MDQWs, and (b) the undoped QWs. The back screens show the laser and absorption spectra.

where  $S_m^{\text{LL}n}$  is the maximum signal emitted from  $\text{LL}n$ , and  $N_{\text{LL}n}$  is the number of photoexcited pairs in  $\text{LL}n$ . If the emission is in direct proportion to the excitation, as we expect from conventional FWM theory, then we should find  $R = 1$ . For the signals shown in Fig. 2 (inset), we find  $R^{\text{undoped}} = 1.3$ , almost as expected, while  $R^{\text{doped}} = 17.5$ , a huge enhancement.

The  $\Delta t$  dependence of the MDQWs signal for this excitation configuration is also rather unusual. According to the RPA theory, the rise time of the  $\Delta t < 0$  signal should be  $1/2$  the decay time for  $\Delta t > 0$ , and this is the measured result for the undoped QWs. This is also the measured result for the signal from LL1 in the MDQWs, as seen in Fig. 2, which shows  $S_{\text{SR}}^{\text{doped}}(\Delta t, \omega)$  for two values of  $\omega$ , corresponding to the maximum signal from LL0 and LL1.

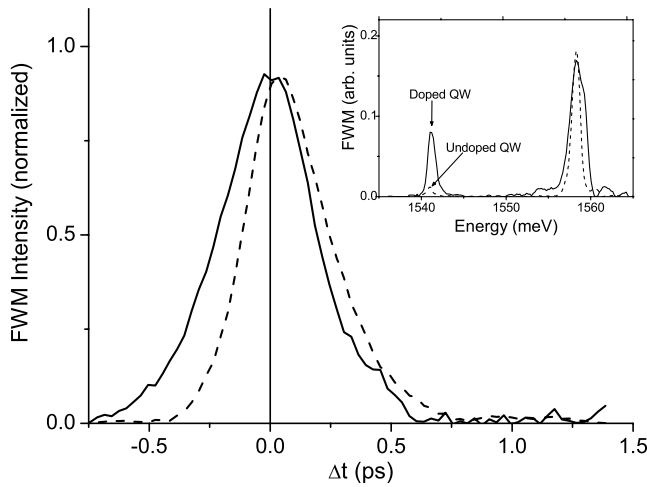


FIG. 2. FWM vs time delay  $\Delta t$  for the MDQWs at  $B = 8$  T. The solid curve is the signal from LL0 and the dashed curve is from LL1. The laser is tuned to excite LL1 (60:1 over LL0), and the signals have been normalized for clarity. The decay time for both curves is  $\approx 0.25$  ps. The rise time for the LL1 signal is 0.13 ps, as expected from mean-field theory, while for LL0 it is 0.27 ps. The inset shows the SR-FWM signal at  $\Delta t = 0$  ps for this excitation from both the doped (solid) and undoped (dashed) QWs.

Surprisingly, the signal from LL0 is almost symmetric as a function of  $\Delta t$  with comparable signals for  $\Delta t < 0$  and  $\Delta t > 0$ . Such a large signal for  $\Delta t < 0$  can only be a result of correlation effects beyond the RPA [6].

We also measured the dependence of  $S_{\text{SR}}^{\text{doped}}(\Delta t, \omega)$  on several additional parameters. By changing the magnetic field, we confirmed that the beat frequency changes with the cyclotron energy and is very close to the LL spacing, and that the LL0 signal is present only for magnetic fields large enough that LL0 is partly empty (filling factor  $\nu < 2$  in the quantum Hall notation). By varying the width of the laser pulse, we determined that the anomalous signal from LL0 requires a small direct excitation of the level. When the pulse is narrowed so that only 1/100 of the carriers are excited into LL0 (rather than the 1/60 in the data discussed above), the LL0 signal drops by a factor of 50. We also measured  $S_{\text{SR}}^{\text{doped}}(\Delta t, \omega)$  as a function of the incident power, varying the photocarrier density in the range  $n/10 \rightarrow n$ . The increase in excitation strength led to large changes in  $S_{\text{SR}}^{\text{doped}}(\Delta t, \omega)$ , especially in the LL0 signal. When the laser was tuned to excite LL1 preferentially, the LL0 signal developed beats as a function of  $\Delta t$ , with a very large minimum at  $\Delta t = 0$ , although the signal remained symmetric about  $\Delta t$  for all powers (see Fig. 2).

Importantly, increasing the power suppressed the relative strength of the LL0 signal, so that the two samples start to look more similar (at  $B = 8$  T,  $R^{\text{doped}}/R^{\text{undoped}} \approx 15 \rightarrow 4$  when the excitation  $\approx n/10 \rightarrow n$ ). This can be understood qualitatively, since as the density of photoexcited carriers approaches that of the 2DEG, the X-2DEG correlations are reduced, and the X-X interactions between excitons begin to dominate.

To understand these results, we calculate the polarization equation of motion by separating the interacting X and 2DEG contributions to the semiconductor wave function in a way that generalizes our treatment of the undoped system [16]. We must include, in addition to the X-X interaction present in undoped QWs, the interaction between the photoexcited X and 2DEG. The inter-LL excitations of the 2DEG, which are important in these experiments, are magnetoplasmons (MP) [11]. In the undoped case, the excited states of the system can be written as products of distinct phonon and X states. However, here both the MP and X excitations are made of electrons, and the exchange effects complicate such a factorization. Therefore, we describe the X-2DEG scattering by introducing a basis of new, correlated excited 2DEG states such as the state Y discussed below.

Several elements are important for understanding the role of the 2DEG in the coherent optical response of MDQWs. (i) Overall coherence requires that the system returns to the ground state after any sequence of processes that result in the destruction of two  $\mathbf{k}_2$  photons and generation of one  $\mathbf{k}_1$  photon and one  $\mathbf{k}_s$  photon. (ii) The MP energy is close to the inter-LL magnetoexciton energy [11], and thus one must account for the resonant, but not instantaneous, MP creation/destruction. The panels in the inset of Fig. 3 illustrate these points. Following the absorption of a photon (a), a LL1 electron scatters to LL0 while a  $\text{LL0} \rightarrow \text{LL1}$  MP is created (b). This intermediate state is a new four-particle excitation, which we call Y, which is nearly resonant with the LL1 X state shown in (a). The subsequent emission of a photon leaves a MP excited (c). The process sketched in (a)  $\rightarrow$  (c) is similar to MP Stokes-Raman scattering [12]. However, after propagation in time, the MP is destroyed by the reverse anti-Stokes Raman scattering, (d)  $\rightarrow$  (f), bringing the system back to the ground state. The full sequence (a)  $\rightarrow$  (f) is similar to the familiar coherent anti-Stokes Raman scattering with phonons [18].

The details of our theory will be published elsewhere, but Fig. 3 shows the result of a model calculation, intended to give a qualitative understanding of the effects of the MP excitations on the FWM signal. The model included only LL0 and LL1 X, the levels excited by the laser, as well as the X + MP shakeup states Y and the MP excitations, and only the nonlinear source terms within the second Born approximation. The matrix elements determining the parameters were not deduced from first principles, but chosen to reproduce the experimental data. If we considered only the excitonic LL coupling at the RPA level without the 2DEG (Fig. 3, dotted curves) [19], no significant  $\text{LL0} \rightarrow \text{LL1}$  transfer of oscillator strength was found, consistent with the undoped QWs data. Accounting for the X-Y scattering (Fig. 3, dashed curves) gave a significant transfer of oscillator strength  $\text{LL1} \rightarrow \text{LL0}$ , showing that one needs this coupling to explain a major experimental result. However, the  $\Delta t$  profile is not satisfactory, since the  $\Delta t < 0$  signal is too small. We know from

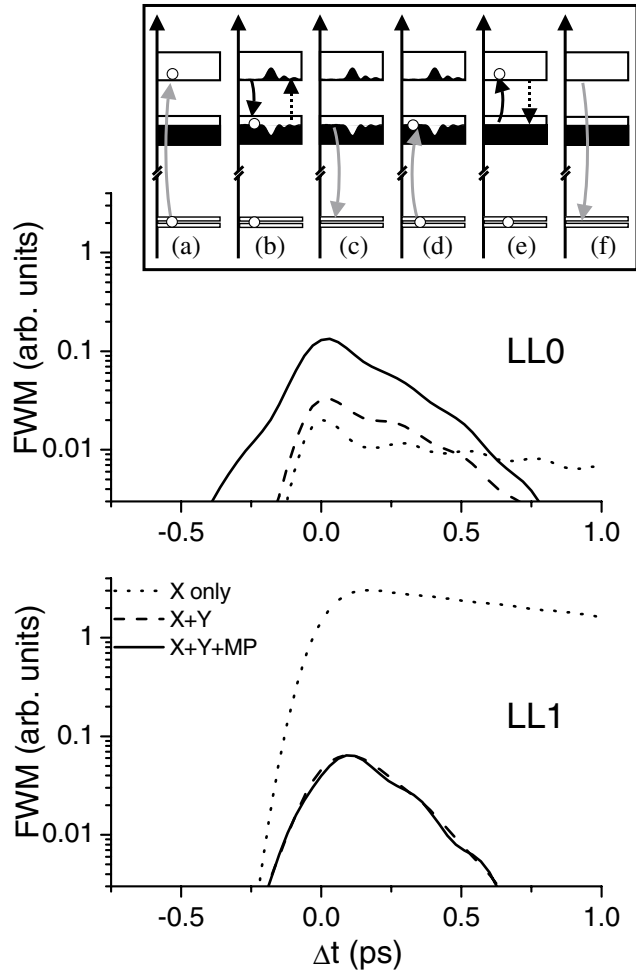


FIG. 3. Model calculation of the FWM profile vs time delay  $\Delta t$ . The inset shows a FWM process which involves the excitation and destruction of a MP. We photoexcite an  $X$  state (a) which scatters with the 2DEG (b) and then is deexcited (c), leaving the 2DEG in an excited state (the VB states are strongly mixed by the magnetic field, allowing this transition [17]). The process is reversed in panels (d)–(f). Note that the excitation shown in (b) does not exist in undoped QWs, and we call this state  $Y$ . The upper graph shows the signal vs  $\Delta t$  at the LL0 energy, and the lower one the signal at the LL1 energy, when only mean-field  $X$ - $X$  interactions are in the model without the 2DEG (dotted line), when we add only the  $X$ - $X$  interactions (dashed line), and when we also add MP scattering processes such as those described in the inset (solid line).

previous studies [6] that a large  $\Delta t < 0$  signal is associated with correlations beyond RPA and non-Markovian processes. It is clear that the MP time propagation [inset, panels (c)  $\rightarrow$  (d)] induces such memory effects. Adding the MP dynamics (Fig. 3, solid curves) increases the  $LL1 \rightarrow LL0$  oscillator strength transfer and enhances the  $\Delta t < 0$  signal, while memory and  $X$ - $Y$  quantum interference effects lead to a more symmetric signal which roughly fits the experimental data.

In conclusion, we have applied coherent wave mixing to investigate the inter-LL ultrafast dynamics of a 2DEG in a

large magnetic field. We observe a large transfer of oscillator strength to the lowest Landau level, and unusual dynamics. These findings are qualitatively explained by a theory that accounts for time-dependent inter-LL scattering mediated by magnetoplasmons.

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